The role of the westerlies and orography in Asian hydroclimate since the late Oligocene

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ADDITIONAL DETAILS ON METHODS AND RESULTS

1. Stable Isotope Geochemistry Method.

In the Tajik Basin, bulk mudstone and fine sandstone samples were collected from the continental redbed in the CD (n=10) and PE (n=62) sections for stable oxygen and carbon isotope analyses. One oyster fossil, namely Ostrea *Kalizky Vialov* (Popov personal communication), was collected from the Bukhara Fm. in the base of the CD section in order to evaluate potential diagenetic processes and primary seasonal $\delta^{18}O$ variability. Soil carbonate nodules were not found in the studied sections, but were collected from the upper Eocene Shurysay member in the ZD section (n=8) and the Miocene Chingou Fm. in the DS (n=3) section for comparison (See Fig. 1 for location). In the western Tarim Basin, 25 samples at an average spacing of 3 m were collected from Paleogene sequences in the UL section.

Thin sections of sandstone samples were examined microscopically to identify regions with fewer than normal authigenic carbonate lithics that can be preferentially drilled for stable isotope analysis. Point counting analysis was performed on each thin section in order to calculate the intergranular volumes (IGV) and the relative proportion of carbonate cements and extrabasinal carbonate lithic (detrital carbonates) within the samples. All calculations are based on ~500 total points counted for each sample. These

27   points were classified as framework grains and interstitial material. The latter was

28   further subdivided into cement, pore space, and matrix. The IGV of a sandstone sample

29   is defined as the volume of interstitial material divided by the total rock volume (Paxton

30   et al., 2002). The extrabasinal carbonate lithics content of a sample is expressed as the

31   ratio of extrabasinal carbonate lithic grains to the carbonate cement count, and the ratio

32   of extrabasinal carbonate grains to total framework grains. Two pilot samples, with

33   relatively high extrabasinal carbonate lithic proportions, were selected from the lower

34   and upper part of the PE section. The hand-block samples were firstly polished and

35   scanned under a Hitachi 3400N Scanning Electron Microscope (SEM) equipped with

36   an Oxford Energy-dispersive X-ray spectroscopy (EDS) system to identify regions of

37   pure carbonate cement, which was then drilled using a dental drill under a binocular

38   microscope.

39   Stable isotope analyses were performed at both the Key Laboratory of Western

40   China's Environmental Systems, Lanzhou University and the Environmental Isotope

41   Laboratory at the University of Arizona. In the Lanzhou University system, a few

42   milligrams of powdered sample were placed in a sealed reaction vessel, which was then

43   flushed with helium and then reacted with 100% phosphoric acid at 72 °C. Carbon

44   dioxide was then sampled from the vessels using a Finnigan Gas-Bench, and stable

45   oxygen and carbon isotope ratios were measured using a Finnigan MAT Delta plus XL

46   mass spectrometer. Repeated analysis of NBS-19 and Merck standards indicates that

47   the laboratory precision of both oxygen and carbon isotope ratios is better than 0.2‰.

48   At the University of Arizona, the $\delta^{18}O$ and $\delta^{13}C$ values of carbonates were measured

49   using an automated carbonate preparation device (KIEL-III) coupled to a gas-ratio mass

50   spectrometer (Finnigan MAT 252). Powdered samples were reacted with dehydrated

51   phosphoric acid under vacuum at 70°C. The isotope ratio measurement is calibrated

52 based on repeated measurements of NBS-19 and NBS-18 and precision is ± 0.1 ‰ for

53 $\delta^{18}O$ and ±0.08‰ for $\delta^{13}C$ (1sigma). $\delta^{13}C$ values of organic matter were measured on

54 a continuous-flow gas-ratio mass spectrometer (Finnigan Delta PlusXL) coupled to an

55 elemental analyzer (Costech) at the Environmental Isotope Laboratory at the University

56 of Arizona. Standardization is based on acetanilide for elemental concentration, NBS-

57 22 and USGS-24 for $\delta^{13}C$, and precision is better than ± 0.10 ‰.

58 **2. Measurement of geochemical elements and calculation of Chemical proxy of**

59 **alteration (CPA).**

60 We focus our effort on fine-grained samples (particle size < 55 μm), because size

61 sorting during transportation and deposition led to some degree of mineralogical

62 differences, which may modify the CPA values. For geochemical analysis, the

63 concentrations of 32 major, trace and rare earth elements/oxides (Cl, S, P, As, Ba, Ce,

64 Co, Cr, Cu, Ga, Hf, La, Mn, Nb, Nd, Ni, Pb, Rb, Sr, Th, Ti, Tl, V, Y, Zn, Zr, Fe_2O_3,

65 SiO_2, Al_2O_3, CaO, Na_2O, K_2O) were determined using a PANalytical PW2403/00 X-

66 ray fluorescence spectrometer. CIA values were calculated using the formula:

67 CPA=$Al_2O_3/(Al_2O_3+ Na_2O)\times100$ (in molar proportions) (Buggle et al., 2011).

68 **3. Clay mineral analysis.**

69 We apply X-ray diffraction (XRD) analysis to identify clay minerals in the

70 sedimentary rocks and scanning electron microscopy (SEM) observations to determine

71 their origin. For XRD analysis, we isolated clay fractions by sedimentation method as

72 described by ref Jackson (2005). Each sample was subdivided into four slides with

73 different treatments, namely Mg-saturation, Mg-saturation in combination with

74 saturation of glycerol, K-saturation, K-saturation combined with heating to 550 °C for

75 1h. Clay slides were scanned on an X-ray diffractometer from 3 to 30° with a step size

76 of 0.02° 2θ/step and speed of 2 s/step, under 45 kV and 30 mA working conditions. The

77 clay minerals were determined by characteristic reflections in XRD patterns (Moore

78 and Reynolds, 1989). For SEM observations, blocks of the selected samples were

79 platinum-coated, and then, were observed on a Hitachi 3400N scanning electron

80 microscope at an accelerating voltage of 13-15 kV. XRD analysis was performed at

81 Lanzhou University and SEM observations were performed both at the university of

82 Arizona and Lanzhou University.

83 **4. Climate model simulation method.**

84 We use an Atmosphere General Circulation Model (AGCM), ECHAM5 (Stepanek

85 and Lohmann, 2012), to evaluate the role of topographic changes in Pamir and Tian

86 Shan in climate during the Late Oligocene. ECHAM5 was adapted for climate research

87 from the weather forecasting model of the European Centre for Medium-Range

88 Weather Forecasts (ECMWF) and developed at the Max Planck Institute in Hamburg

89 (Germany). The model is based on a spectral dynamical core and simulates the

90 troposphere and the lower stratosphere up to a pressure level of 10 hPa. The vertical

91 dimension is organized on a hybrid sigma-/pressure-level system. In our model setup,

92 we use ECHAM5 in T106/L31 resolution (i.e. there are 31 levels present and triangular

93 truncation of the series of spherical harmonics is performed at wave number 106).

94 We design the Pre-industrial (EXP.PI) and the Late Oligocene (EXP.LO)

95 experimental groups to portray the annual precipitation and its $\delta^{18}O$ over Central Asia.

96 The PI experimental group using boundary conditions of pre-industrial period

97 (Stepanek and Lohmann, 2012), whereas the EXP.LO experimental group with

98 additional change in the land-sea distribution in Euroasia (i.e. large area extent of the

99 Paratethys during Late Oligocene) (Fig. S6, Table S3), following the dataset from

100   Markwick (2007), which is overall agreement but slightly larger than paleogeographic

101   reconstructions (Popov et al., 2004; Bosboom et al., 2017).

102   To better understand the impact of tectonic uplift on climate changes over central

103   Asia and constraining the threshold altitude of the mountains that could block westerlies

104   moisture, we conducted sensitivity simulations with progressive height reductions in

105   the Pamir-Tian Shan convergence zone (64ºE-78 ºE, 34.5 ºN-43.5 ºN) for the EXP.PI

106   and EXP.LO experimental group, respectively (Fig. S6, Table S3).

107   **5. Stable Carbon isotope records and its interpretations**

108   **5.1 Results**

109   Carbon isotope compositions of nonmarine strata ($\delta^{13}C_{cc}$) in the Tajik Basin range

110   from -4.59 to 0.22‰, whereas, the coexisting organic matter has $\delta^{13}C$ values ($\delta^{13}C_{org}$)

111   that fall between -27.05 and -24.19‰ (Fig. S7). Both records yield a slightly stepwise

112   increasing trend up-section. $\delta^{13}C_{cc}$ values of carbonate nodule samples from the upper

113   Eocene strata in the adjacent Zadi (ZD) section and the upper Oligocene to lower

114   Miocene strata in the Dashtijum section (DS) averaged at -6.18 and -4.33‰ and are

115   systematically lower than those of the cements samples (Fig. S7).

116   **5.2 Evaluating the $\delta^{13}C_{cc}$ records and its paleoclimate implication**

117   Shallow surface water or groundwater bicarbonate will control the $\delta^{13}C_{cc}$ values of

118   fluvial cements; and it may be derived from the dissolution of limestone or a mixture

119   of other sources. In contrast, the carbon isotope ratios of paleosol carbonate and organic

120   matter are widely used as proxy indicator for paleoecology and paleoclimate changes

121   in terrestrial depositional systems (Cerling et al., 1989). Paleosol $\delta^{13}C_{cc}$ values are

122   controlled by the isotopic composition of soil-respired CO_2, which depends mainly on

123   the photosynthetic pathway of the overlying plants, and on climatic changes where the

124   soil respiration rates are high (Cerling, 1984). Organic matter in soils and sediments

125 has a carbon isotopic composition derived from local plant matter, which also depends

126 on photosynthetic pathway. C3, C4, and crassulacean acid metabolism (CAM) plants

127 exhibit different carbon isotopic compositions, ranging from -22‰～-30‰, -9‰～-

128 19‰, and -14~-34‰ (O'Leary, 1988), respectively. For example, paleosol carbonates

129 precipitated in equilibrium with CO_2 derived solely from C3 plants have $\delta^{13}C_c$ values

130 ranging from -9 to -13‰, and are associated with organic matter that have $\delta^{13}C_{org}$ values

131 typically $\leq -24‰$ (Cerling, 1984).

132     $\delta^{13}C_{org}$ values in the Tajik Basin fall between -27.05 and -24.19‰ (Fig. S7),

133 indicating that the overlying vegetation type was made entirely by C3 plants. It is

134 important to note, however, that the carbon isotope ratio difference between cements

135 and associated organic matter is much larger than the expected difference for soil

136 carbon isotope ratios. Instead of the typical 14‰ to 16‰ difference, cements and

137 coexisting organic matter have $\delta^{13}C$ values that differ by 22.5 to 25.6 ‰. This suggests

138 that the $\delta^{13}C_{cc}$ values obtained from cements in this study are strongly affected by

139 groundwater DIC rather than CO_2 derived from the soil. The $\delta^{13}C_{cc}$ values of cements

140 in this study, therefore, have little to do with the regional climate.

141   The $\delta^{13}C$ values of soil carbonate nodule samples from upper Eocene strata in the

142 ZD section and the upper Oligocene to lower Miocene strata in the DS section have

143 average values between -6.18 and -4.33‰, which are systematically higher than the

144 typical $\delta^{13}C$ values from a pure C3 source (c.a. -11‰). These surprisingly high carbon

145 isotope ratios, commonly observed in arid central Asia, may have arisen from very low

146 soil-respiration-rates due to limited primary productivity (Caves et al., 2016). This leads

147 to elevated $\delta^{13}C_{cc}$ values because of mixing with the atmospheric CO_2 and perhaps the

148 water-stress effect on plant with effects on $\delta^{13}C$ values (Caves et al., 2016). Because

149 both of these factors are caused by arid climate conditions, we infer that an arid climate

150   had been established in the Tajik Basin since at least the late Eocene.

151   **ADDITIONAL REFERENCES CITED**

152   Cerling, T., Quade, J., Wang, Y., and Bowman, J., 1989, Carbon isotopes in soils and
153   palaeosols as ecology and palaeoecology indicators: Nature, v. 341, no. 6238, p.
154   138-139.
155   Cerling, T. E., 1984, The stable isotopic composition of modern soil carbonate and its
156   relationship to climate: Earth and Planetary Science Letters, v. 71, no. 2, p. 229-
157   240.
158   Jackson, M. L., 2005, Soil chemical analysis: advanced course, UW-Madison Libraries
159   Parallel Press.
160   Moore, D. M., and Reynolds, R. C., 1989, X-ray Diffraction and the Identification and
161   Analysis of Clay Minerals, Oxford university press Oxford.
162   O'Leary, M. H., 1988, Carbon isotopes in photosynthesis: Bioscience, v. 38, no. 5, p.
163   328-336.
164   Berger, A., 1978, Long-Term Variations of Daily Insolation and Quaternary Climatic
165   Changes: Journal of the Atmospheric Sciences, v. 35, no. 12, p. 2362-2367.
166   Wei, W., and Lohmann, G., 2012, Simulated Atlantic Multidecadal Oscillation during
167   the Holocene: Journal of Climate, v. 25, no. 20, p. 6989-7002.
168   Zhang, X., Lohmann, G., Knorr, G., and Xu, X., 2013, Different ocean states and
169   transient characteristics in Last Glacial Maximum simulations and implications
170   for deglaciation: Clim. Past, v. 9, no. 5, p. 2319-2333.
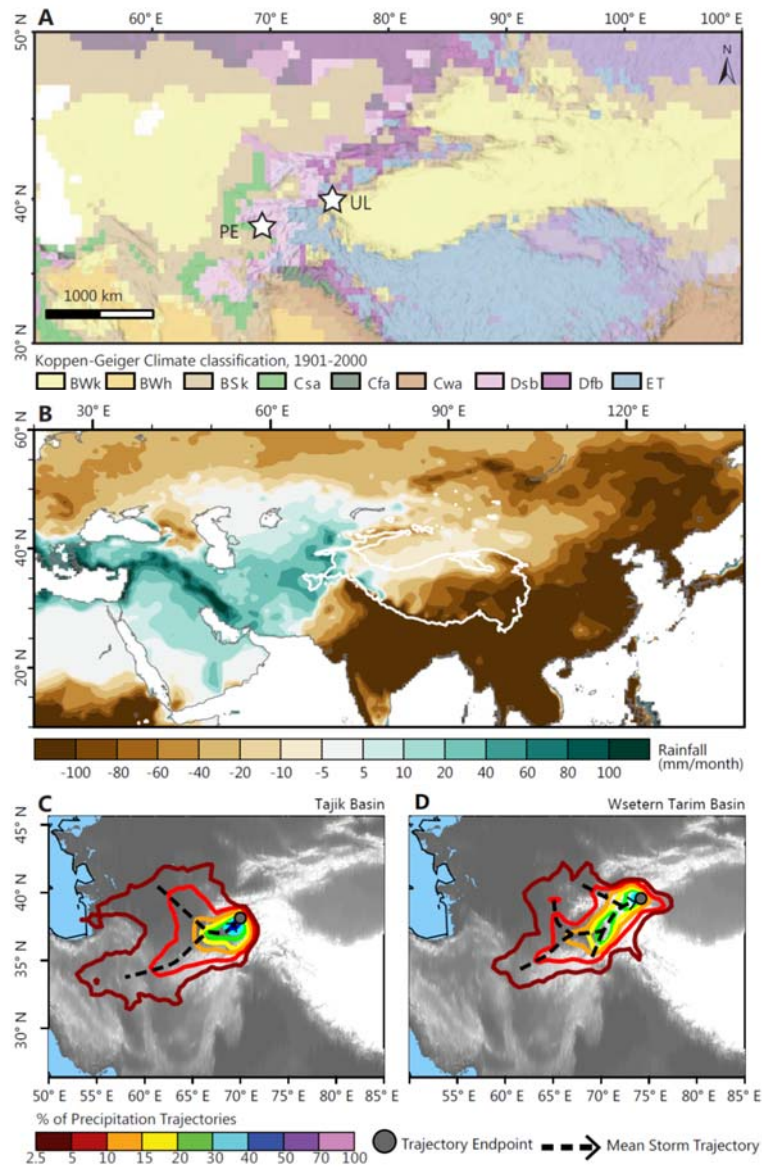
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SUPPLEMENTAL FIGURES

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Fig. S1. Climatic setting over central Asia. (**A**) Koppen-Geiger climate classification of the mid-latitude Euro-Asia (https://www.arcgis.com), showing the west-east environment differences over central Asia. (**b**) Seasonal precipitation differences between boreal winter (DJF) and summer seasons (JJA) based on the NCEP/NCAR atmospheric reanalysis data (Kalnay et al., 1996) during 1971-2000. (**C-D**) Contours of precipitation-producing storms over the Tajik and the western Tarim basins. Mapped using the Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT) (Caves et al., 2015). Note that nearly all precipitation-producing storms originate to the west, indicating the westerlies moisture plays a leading role in controlling hydroclimate changes in the working sections.

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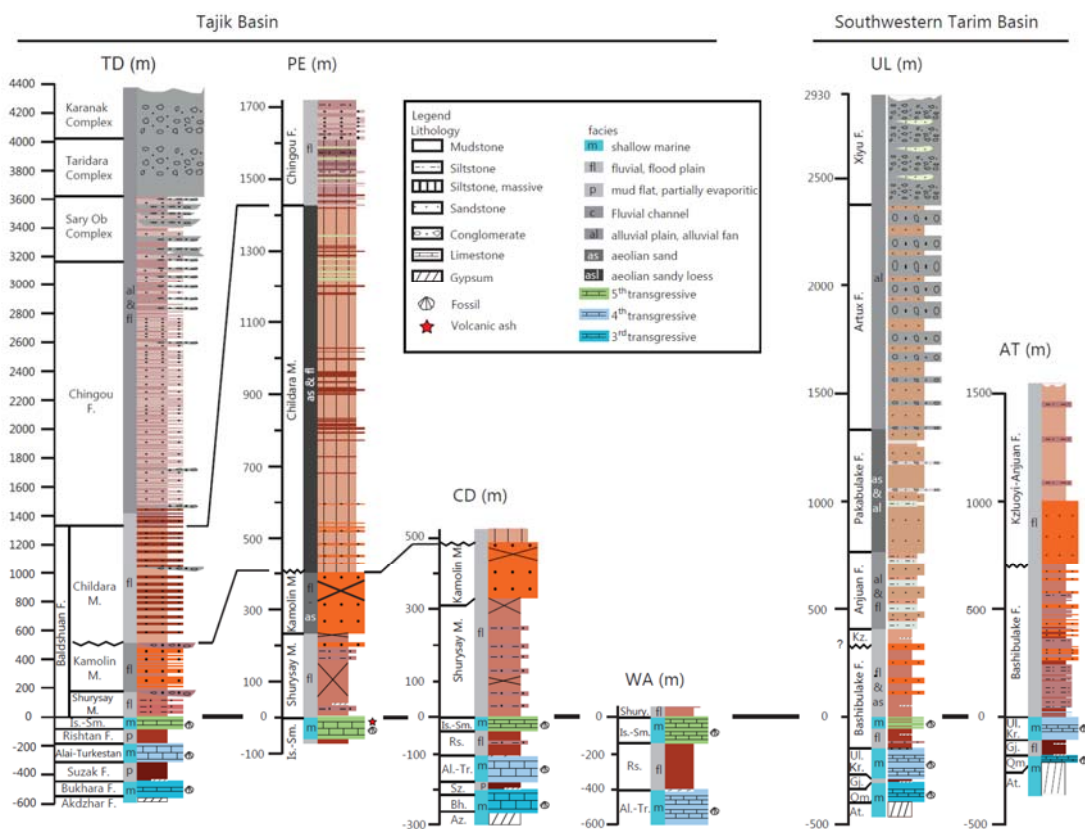
Fig. S2. Stratigraphy of the Cenozoic sequences in the Tajik and the western Tarim basins (after Wang et al., 2019 and references there in). The correlation is based on lithology, paleontology, and regional stratigraphic subdivisions. Zero level is defined by the top of the stratigraphically highest shallow marine strata for comparing purpose. Abbreviations for formation/member name: Az.-Akdzha; Bh.-Bukhara; Sz.-Suzak; Al.-Alai; Tr.-Turkestan; Rs.-Rishtan; Is.-Isfara; Sm.-Sumsar; Shury.-Shurysay; At.-Aertashi; Qm.-Qimugen; Gj.-Gaijitage; Kr.-Karatar; Ul.-Ulagen; Kz.-Keziluoyi.

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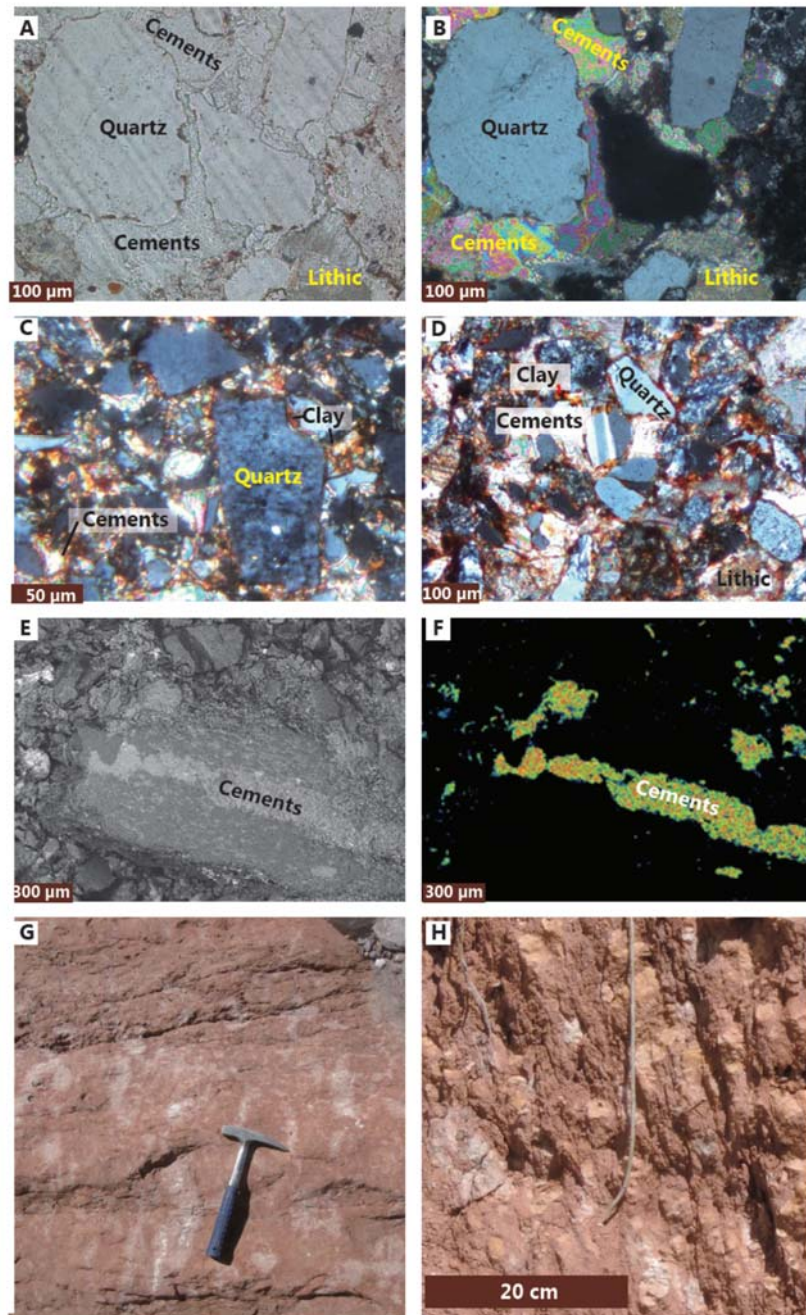
Fig. S3. Petrographic images showing the compositions of carbonate materials.
Thin section photography of a sandstone samples under PPL (**A**) and XPL (**B**), showing both carbonate cements and lithics preserved in the samples. (**C-D**) Micromorphology of silt stone samples from the PE section, showing weakly-developed pedogenic features such as illuvial clay (**C**) and calcite crystallite b-fabric (**D**) within the samples. (**E-F**) Scanning Electron Microscope (SEM) image (**E**) and Energy-dispersive X-ray spectroscopy (EDS) showing distribution of element Ca surrounding a quartz, indicating pure carbonate cements which were drilled for stable isotope analysis. (**G-H**), photography of shows paleosol carbonate nodule in the Miocene Chingou Fm. from the DS section (**G**) and the Upper Eocene Shurysay Member in the CD section (**H**).

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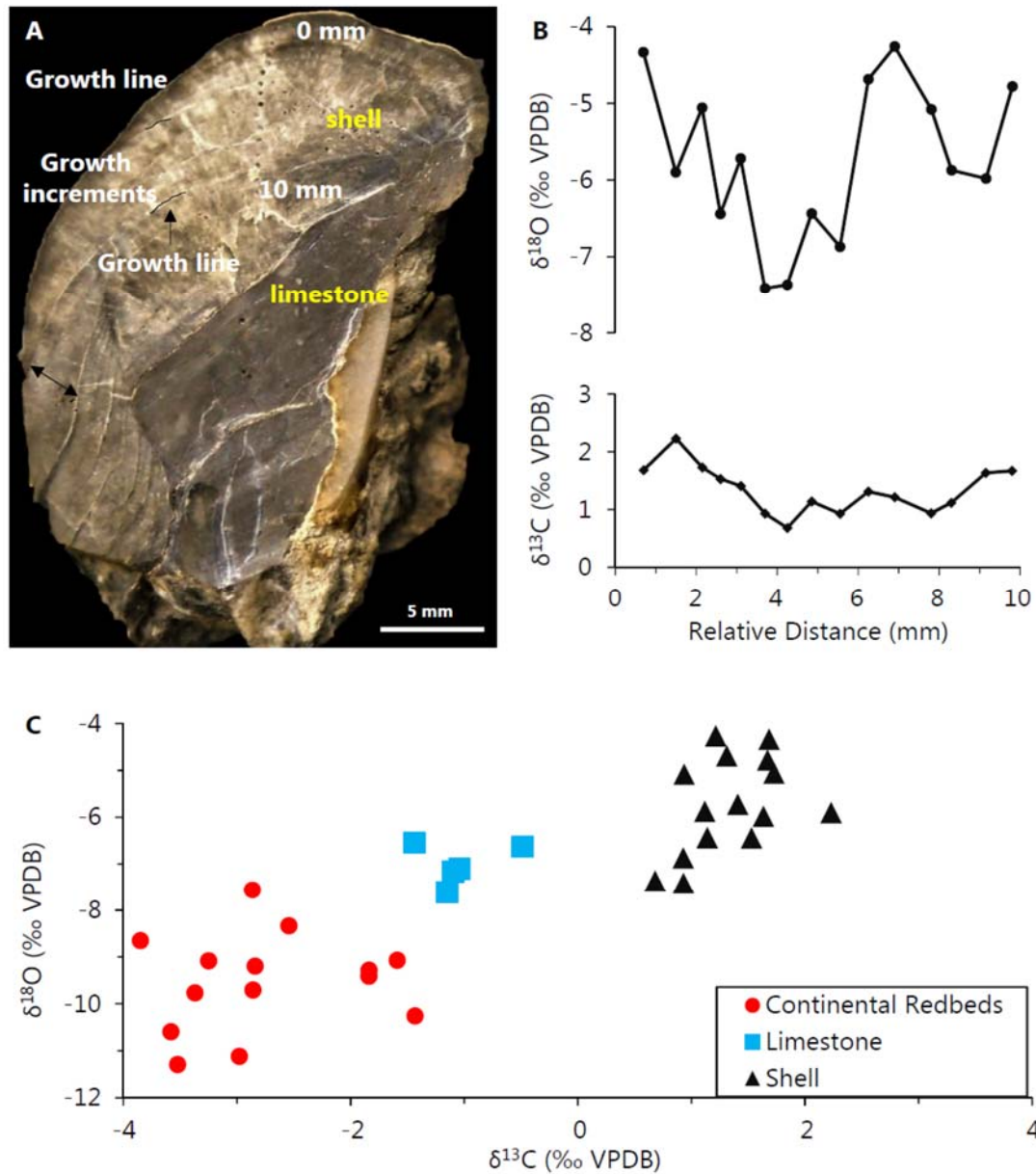
Fig. S4. High-resolution δ¹⁸O and δ¹³C of an Oyster fossil. (**A**) Polished cross sections of Ostrea *kalizky Vialov* revealing annual growth bands. (**B**) high-resolution δ¹⁸O and δ¹³C values along a growth line of the shell. Distance is measured from the first drilled micro-sample at the edge of the shell. (**C**) Cross plot of δ¹⁸O versus δ¹³C for the Oyster fossils, limestone, and adjacent continental red beds in the lower part of the studied sections. The differences between different type of sediments indicate limited diagenetic alteration.
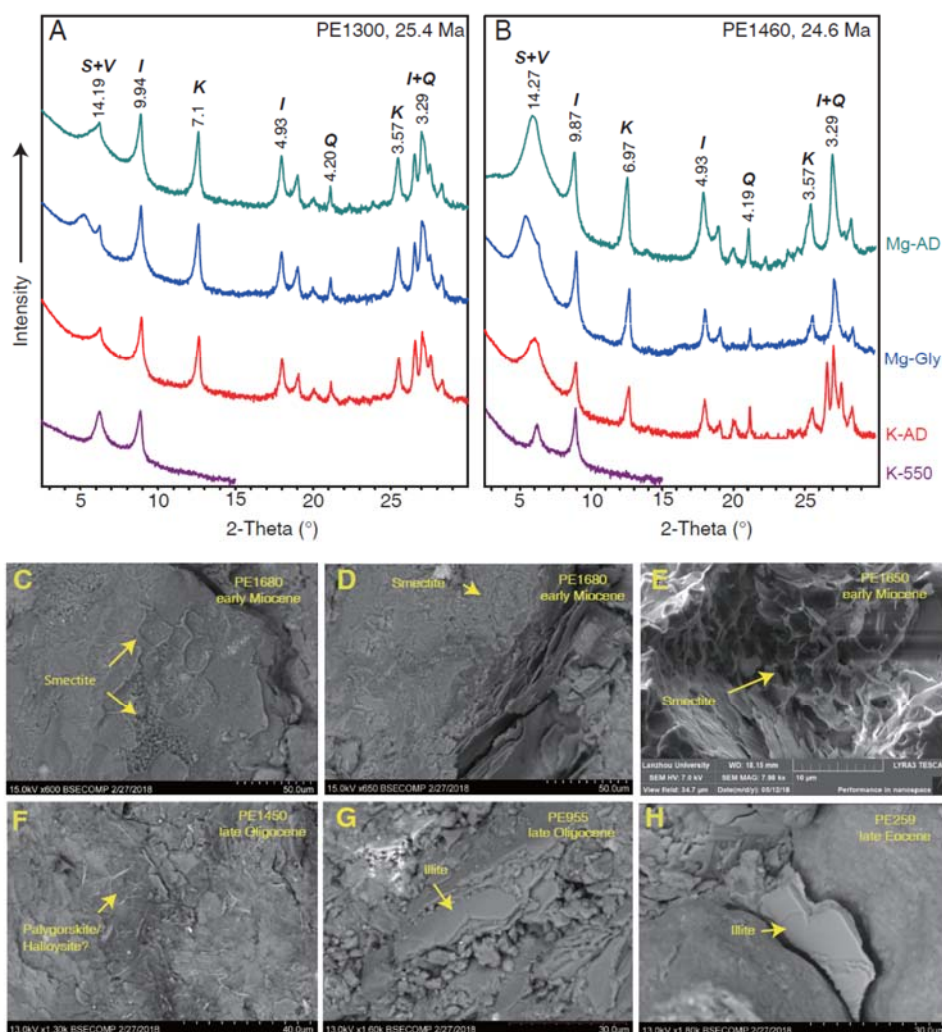
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Fig. S5. Clay mineral records. (A-B) Example of multi-treatment X-ray diffraction (XRD) patterns of representative sample from the PE section. S: smectite, I: illite, K: kaolinite, V: vermiculite; Ch: chlorite, Q: quartz, Mg-AD: magnesium saturated and air-dried; Mg-Gly: magnesium and glycerol saturated; K-AD: potassium saturated and air-dried; K-550: potassium saturated and 550 °C heating. **(C-H)** SEM image of clay minerals.
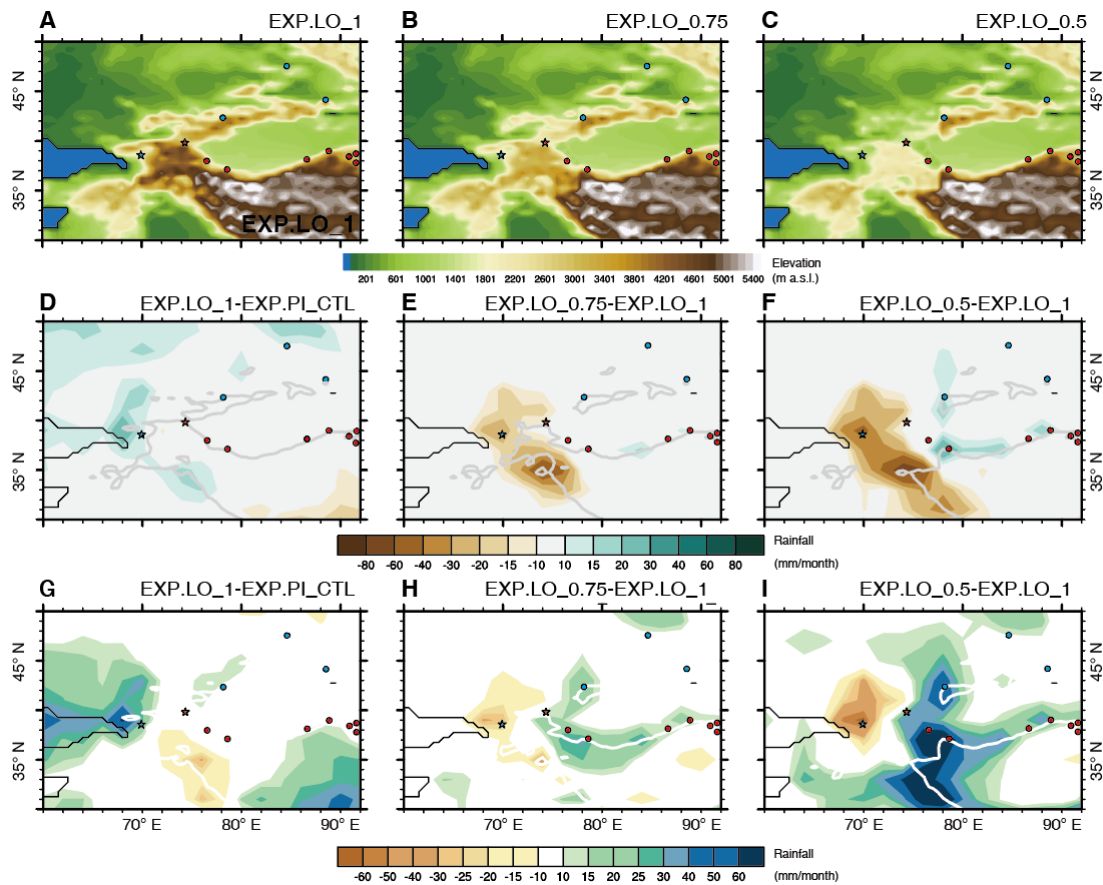
Fig. S6. Climatic simulations. (**A-C**) Topographies and ocean bathymetries designed for high mountains (100% relief, ~4000 m), medium mountain (75% relief, ~3000 m), and low mountain (50% relief, ~2000 m) scenarios. (**D**) Annual precipitation differences between Late Oligocene high mountains experiment and pre-industrial control experiment, showing the Late Eocene climate in central Asia is wetter than the pre-industrial period due to the presence of the Paratethys. (**E-F**) Annual precipitation differences between Late Oligocene medium (low) mountain and high mountains experiment, revealing decreases precipitation in the mountain regions and its western front, and increases precipitation in the eastern side associated with reducing topography in the Pamir-Tian Shan convergence zone. (**G**) Seasonal precipitation differences between Late Oligocene high mountains experiment and pre-industrial control experiment. (**H-I**) Seasonal precipitation differed between Late Oligocene medium (low) mountain and high mountains experiment, suggesting that the east-west seasonal precipitation differences become much weaker associate reduction of elevations in the Pamir-Tian Shan convergence zone.
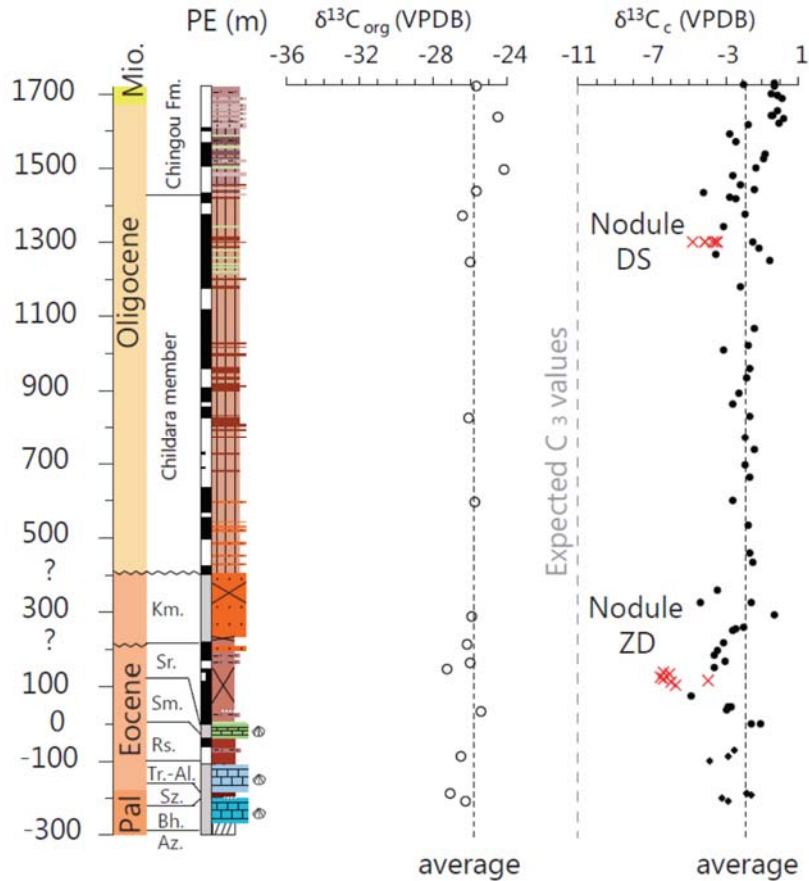
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Fig. S7. Stable carbonate isotope of the early Cenozoic sequences in the Tajik Basin. The $\delta^{13}C$ values of paleosol carbonate from the DS and ZD sections are indicated by red cross for comparison. $\delta^{13}C$ values of soil carbonate nodule samples have average values between -6.18 and -4.33‰, which are systematically higher than the typical $\delta^{13}C$ values from a pure C3 source (c.a. -11‰), which may have arisen from very low soil-respiration-rates due to limited primary productivity, which are caused by arid climate conditions. Note that the carbon isotope ratio difference between cements and associated organic matter is much larger than the expected difference (~14‰ to 16‰) for soil carbon isotope ratios. This suggests that the $\delta^{13}C_{cc}$ values obtained from cements in this study are strongly affected by groundwater dissolved inorganic carbon (DIC) rather than CO_2 derived from the soil. The $\delta^{13}C_{cc}$ values of cements in this study thus have little to do with the regional climate.

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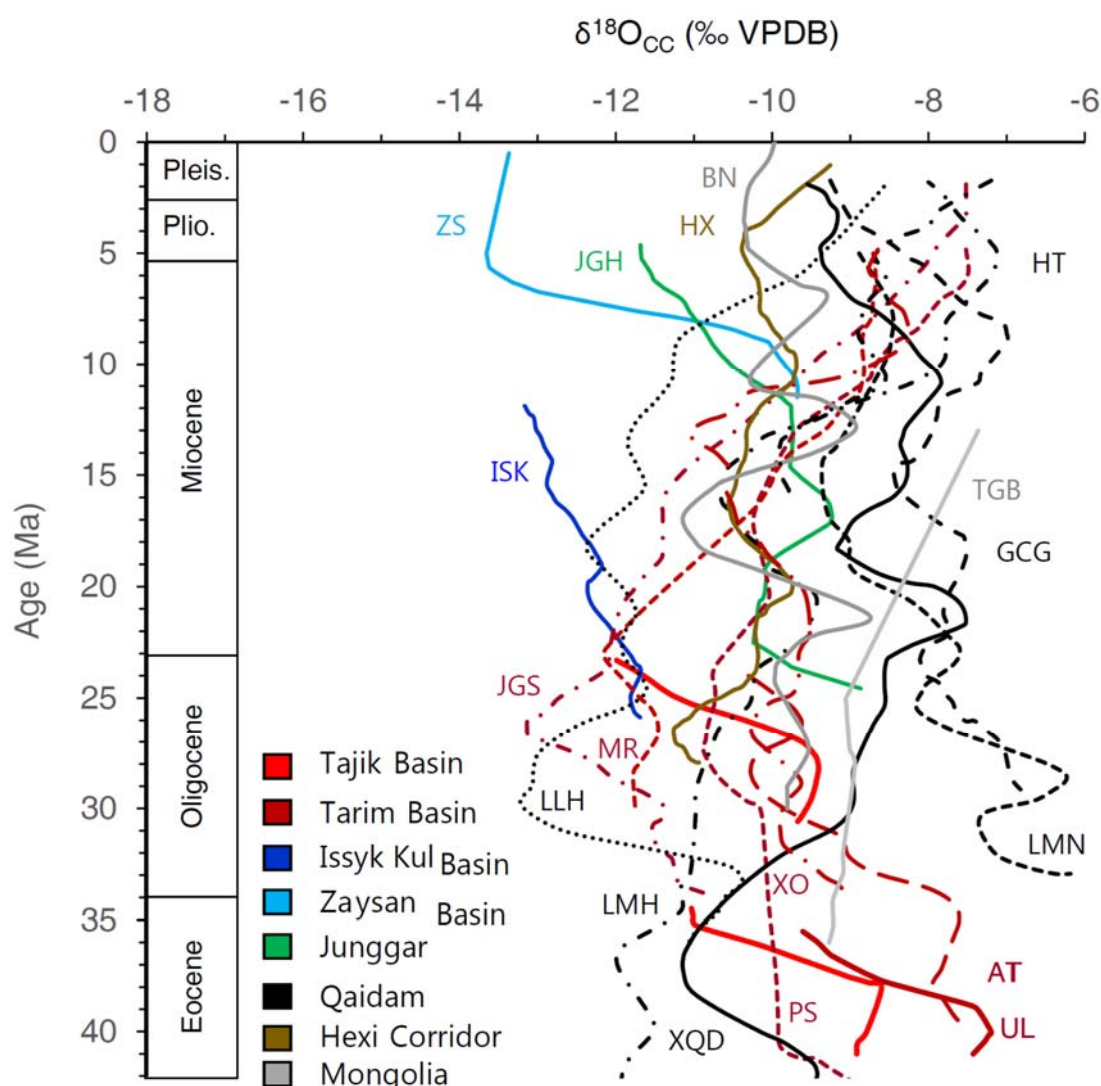
Fig. S8. Citations of δ¹⁸O$_{CC}$ records. Data from each section were smoothed using
an Epanechnikov kernel with a 1 Ma bandwidth. Colored lines correspond to different
basins. ISK-Issyk Kul (Macaulay et al., 2016), ZS-Zaysan (Caves et al., 2017), JGH-
Jingouhe (Charreau et al., 2012); HT-Huaitoutala (Zhuang et al., 2011); BN= Biger
Noor, TGB-Taatsin Gol (Caves et al., 2014); AT-Aertashi, PS-Puska, MR-Miran River,
JGS-Janggalsay, XO-Xorkol, HX- Hexi Corridor, GCG-Ganchaigou, LLH-Lulehe,
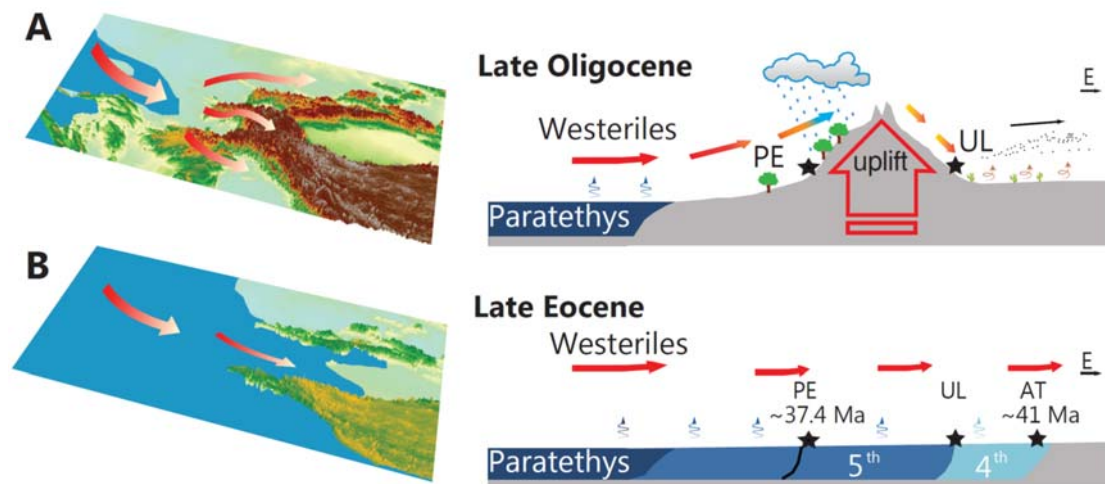LMH-Lake Mahai, LMN-Laomangnai, XQD- Xiao Qaidam (Kent-Corson et al., 2009).

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Fig. S9. Schematic models showing topographic-atmospheric configuration changes over central Asia during the Cenozoic.

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SUPPLEMENTAL TABLE CAPTIONS

261 **Table S1**. **Stable isotope data** (provided as excel format).

262 **Table S2. CPA data**, calculated from fine-grained samples, in the Tajik Basin

263 (provided as excel format).

264 **Table S3. Boundary conditions in climate simulations.** The topography of the
265 Pamir-Tian Shan convergence zone (64ºE-78 ºE, 34.5 ºN-43.5 ºN) was expressed as a
266 percentage to modern elevation and approximate height. Land-sea distribution during
267 the late Oligocene was cited from Markwick (2007). Orbital parameters are calculated
268 according to Berger (1978). The pre-industrial (PI) climate boundary conditions are set
269 from a PI simulation, which has been run into equilibrium over several thousand years
270 (Wei et al., 2012; Zhang et al., 2013).
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Experiment ID	Topography	Land-sea	Orbital	CO_2 (ppm)	CH_4 (ppb)	N_2O (ppb)	SST Ice volume Sea level
EXP.LO_1	100% / 4000m	Late Oligocene	PI	280	760	270	PI
EXP.LO_2	75% / 3000m	Late Oligocene	PI	280	760	270	PI
EXP.LO_3	50% / 2000m	Late Oligocene	PI	280	760	270	PI

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Table S4. Sandstone point counting results.

Stratigraphic Position (m)	Total Counts	Extra-basinal	Cement	Matrix	Pore	IGV (%)	Extralbasinal/ Cement (%)	Extralbasinal /Total (%)
PE250	500	15	96	33	4	26.60	15.63	3.00
PE387	497	8	28	64	7	19.92	28.57	1.61
PE425	500	19	49	75	1	25.00	38.78	3.80
PE1528	500	13	52	52	4	21.60	25.00	2.60
PE1614	500	25	57	118	22	39.40	43.86	5.00
PE1680	500	21	56	71	14	28.20	37.50	4.20
UL63	499	7	42	34	58	26.85	16.67	1.40
UL245	499	3	20	40	55	23.05	15.00	0.60
UL851	499	1	88	19	12	23.85	1.14	0.20
UL1095	500	8	107	17	25	29.80	7.48	1.60
UL1230	500	3	73	22	63	31.60	4.11	0.60